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**Probing and Manipulation of Coherent States in Ferromagnetic  
Narrow Gap Semiconductors with an Eye Towards Developing  
Concepts for New Device Functionalities**

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**Final Report**

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## **Status of efforts:**

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The goal of this project has been to study coherent phenomena in narrow gap semiconductors (NGSs) including III-V ferromagnetic structures, such as InMnAs and InMnSb, with an emphasis on dynamical aspects, such as charge and spin dynamics. In light of the growing interest in spin-related phenomena and devices, there is now renewed interest in the science and engineering of NGSs and our results are important for understanding the electronic and magnetic states in these material systems. Narrow gap semiconductors have significant potentials for applications in infrared *spin photonics* and in spin transport devices due to small energy gap, and much higher carrier mobility than in other III–Mn–V semiconductors.

I have developed strong collaborations with several materials scientists and user facilities. Our recent observations have been reported in new publications and several talks and poster presentations. Our activities will advance the following AFOSR missions:

- a.** Fundamental understanding of materials that change their physical characteristics in response to an external stimulus;
- b.** Multifunctional materials;
- c.** Fundamental research of electronic, photonic, and magnetic properties of solid state materials, structures, and devices;
- d.** Developing concepts for advanced devices.

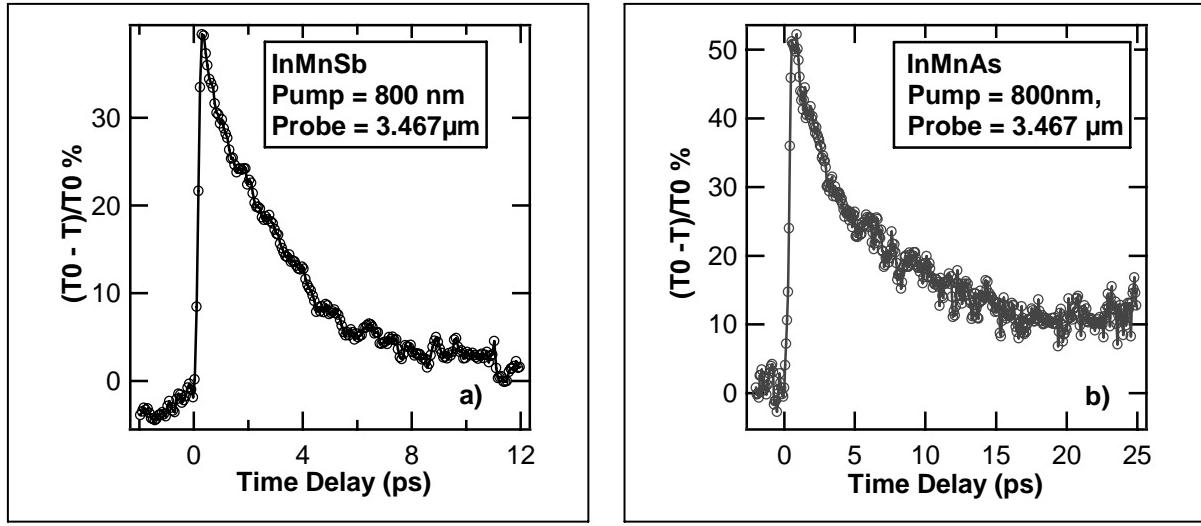
## **Accomplishments/ New Findings:**

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After the last year report my group pursued several new experiments and the examples are presented in this final report. The publications on the basis of the recent observations are under preparation. Several time resolved differential transmission (TRDT) schemes were employed to provide insight into the time scales and the nature of microscopic interactions in metalorganic vapor phase epitaxy (MOVPE) grown InMnAs and InMnSb. The MOVPE grown InMnAs structure, was an 800 nm thick film with a Mn content of ~ 4 %, a hole concentration  $p=1.35 \times 10^{18} \text{ cm}^{-3}$ , and a mobility of  $142 \text{ cm}^2/\text{Vs}$ . The 200 nm thick InMnSb film has a hole concentration  $p=3 \times 10^{19} \text{ cm}^{-3}$ , and a mobility of ~ $198 \text{ cm}^2/\text{Vs}$  with the Mn content of 3.7 %.

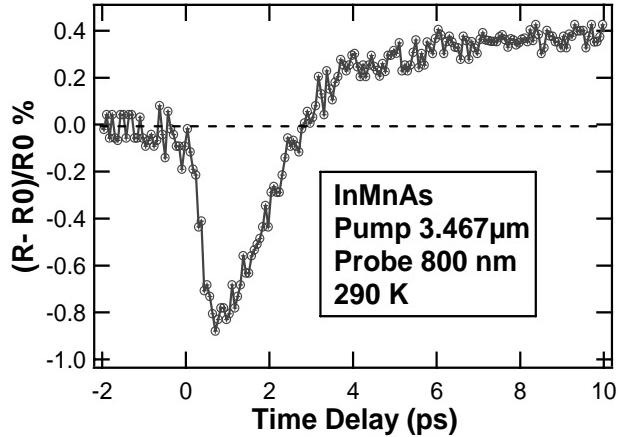
Most of the understanding of the carrier relaxation in the MBE grown narrow gap (III,Mn)V ferromagnetic structures has been on the basis of two color differential reflectivity spectroscopy with pump and probe pulses ranging from  $1.2\text{--}2\mu\text{m}$  and  $650\text{--}850 \text{ nm}$ , respectively [1]. The information extracted from the differential reflectivity can be affected by multi-reflections in the multi-layer structures. In this work, our TRDT measurements demonstrate complex and tunable dynamics of the photo-excited carriers in these ferromagnetic films which have not been reported before.

Figure 1 demonstrates the examples of two-color TRDT, where the pump and probe pulses were tuned at 800 nm and 3.467 m, respectively. The TRDT in these two samples demonstrate different relaxation patterns. The differential transmission change, in InMnSb is about 15%; whereas, for InMnAs under the same experimental conditions, the change exceeds 50%. In addition, the fast component of the carrier relaxation in the InMnSb film is rather different compared to the InMnAs and the photo-induce carriers fully relax in the time scale of 12 ps. In both samples 800 nm pump pulses can generate electrons that are energetic to have the possibility scattering between the X, L, and  $\Gamma$  valleys in the conduction band [2-3]. Our earlier degenerate MIR pump/probe measurements demonstrated different relaxation dynamic possibly influenced by non-linear effects involved in a degenerate scheme [4-5].



**Fig. 1:** Two-color differential transmission measurements in ferromagnetic films, at 290 K. The pump/probe pulses were 800 nm and 3.467  $\mu$ m, respectively. In case of InMnSb, the photo-induced carriers were relaxed in the time scale of 12 ps; whereas, in the InMnAs the carriers were not fully relaxed in a time scale longer than 25 ps.

As shown in Fig. 2, when the order of the pump and probe pulses were reversed and tuned to 3.467  $\mu$ m and 800 nm, respectively, the differential reflectivity as a function of time delay in the InMnAs was measured. The initial sharp decrease in the differential reflectivity can result from free carrier Drude absorption whereas; the alteration of the dielectric function of the film through changes in the electron and hole distribution functions, can be responsible for the observed sign change of the differential reflectivity. A similar fast carrier dynamics and the sign change in the transient reflectivity have been reported in MBE grown InMnAs and InAs films <sup>[1,6]</sup>. The possibility of increasing the relaxation time of the photo-excited carriers by injection carrier above the gap into the satellite valley can be important in developing devices on the basis of these ferromagnetic materials.



**Fig. 2:** Two-color normalized transient reflectivity in ferromagnetic films, at 290 K. The pump/probe orders have been reversed compared to Fig. 2. A similar fast carrier dynamics and the sign change in the transient reflectivity have been reported in MBE grown InMnAs. The alteration of the dielectric function of the film through changes in the electron and hole distribution functions, can be responsible for the sign change.

In our measurements, the MIR laser source was a difference frequency generator (DFG), which mixes the signal and idler beams from an optical parametric amplifier (OPA). The OPA itself was pumped by an amplified Ti:sapphire oscillator with a repetition rate of 1 KHz. The pulses had a duration of  $\sim 100$  fs defining the resolution of the measurements. The differential transmissivity as a function of the time delay between the pump and probe pulses, using a liquid N<sub>2</sub>-cooled MCT detector was measured. Similarly, differential transmission measurements can extract the carrier relaxation time.

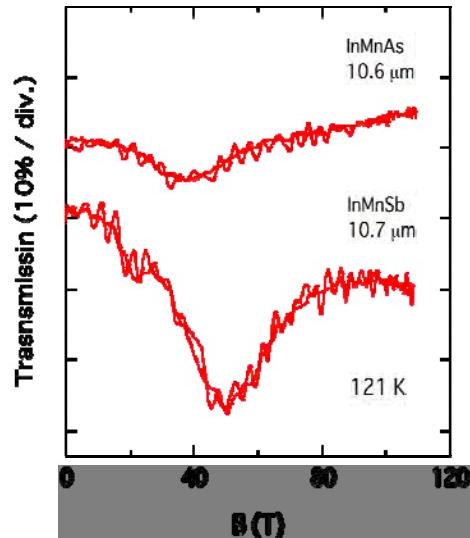
In addition to work at Virginia Tech, my group has been collaborating with large-scale facilities, at two national labs (The National High Magnetic Field Labs in Florida and Institute for Solid State Physics in Japan), to provide information on the properties of ferromagnetic material systems. An example of these collaborative efforts is presented here.

### Cyclotron Resonance at High Magnetic Fields

The carrier-mediated ferromagnetism in magnetic III-V semiconductors has provided a novel system to study the physics of itinerant carriers interacting with localized spins of the magnetic impurities. The nature of the carriers mediating ferromagnetism has been an open question. This fact motivated us to probe Cyclotron Resonance (CR) in the ferromagnetic structures to understand the nature of the carrier states. CR is a direct and accurate method for determining the effective mass and therefore the energy dispersion of carriers. In DC transport measurements it is difficult to extract the contributions of the mass and scattering time independently. Since these magnetic semiconductors usually have low carrier mobilities, a very high magnetic field is necessary to observe cyclotron resonance (CR) (i.e.,  $\omega_c\tau > 1$ )

Ferromagnetic semiconductors are important materials for development of spintronic devices. While effort in this area was made primarily on GaMnAs, other ferromagnetic III-Mn-V alloys have also been developed, including the narrow gap ferromagnetic alloys InMnAs and InMnSb. For example, important advances have now been made in the MOVPE growth of these latter systems [7-8]. Investigation of the electronic structure of III-Mn-V alloys by techniques such as the cyclotron resonance (CR) can shed important light on the origin of ferromagnetism and the *p-d* exchange interaction in III-Mn-V systems. In this work we report on CR experiments carried out on the ferromagnetic InMnAs and InMnSb films, on which clear resonance signals have been successfully observed in high magnetic fields.

In<sub>1-x</sub>Mn<sub>x</sub>As films were grown by MOVPE at the Northwestern University and In<sub>1-x</sub>Mn<sub>x</sub>Sb films at the University of Notre Dame by low temperature MBE [9]. The Mn content (*x*), thickness (*d*), and hole concentration (*p*) are *x* $\sim 0.02$ , *d*=200 nm, *p* $\sim 2 \times 10^{18}$  cm<sup>-3</sup> for In<sub>1-x</sub>Mn<sub>x</sub>As, and *x* $\sim 0.02$ , *d*=230 nm, *p* $\sim 1 \times 10^{20}$  cm<sup>-3</sup> for In<sub>1-x</sub>Mn<sub>x</sub>Sb. The *T<sub>c</sub>*'s are about 320 K and 10 K in In<sub>1-x</sub>Mn<sub>x</sub>As and In<sub>1-x</sub>Mn<sub>x</sub>Sb, respectively. CR measurements were performed using CO<sub>2</sub> and H<sub>2</sub>O lasers, providing laser radiation at 10.6, 10.7, and 16.9  $\mu$ m wavelengths. Magnetic fields exceeding 100 T were generated by a single turn coil technique.



**Fig. 3:** Cyclotron resonance spectra of InMnAs and InMnSb.

Figure 3 shows the CR spectra at 121 K. The effective masses deduced from these resonance peak positions are  $0.037m_0$  for InMnAs and  $0.051m_0$  for InMnSb. The observed effective mass in InMnAs is consistent with the heavy hole (HH) mass reported in an earlier CR study of *p*-type InMnAs with  $T_c$  of ~10 K [10]. This HH effective mass is quite different from the classical HH band edge mass in InAs ( $0.35m_0$ ) because of the known quantum effect at high magnetic fields with for small Landau level indices ( $n = 0,1,2,..$ ) [10]. The CR in ferromagnetic InMnSb was observed for the first time. The deduced mass is also considerably smaller than the band edge HH mass of InSb ( $0.32m_0$ ), similar to the result obtained for InMnAs. Detailed theoretical calculations will provide information on the carrier states in the valence band of these ferromagnetic alloy systems.

### **Personnel Supported**

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Graduate Students supported: Matt Frazier (Currently a Post Doctoral associate at the Physics Department, University of Maryland), Mithun Bhowmick, Travis Merritt. In addition, three undergraduate students (Thomas How, Jonathan Cates and Jose Umanzor-Alvarez) were partially supported.

### **Publications Resulted from AFOSR Support**

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- 1-** M. Frazier, J. G. Cates, J. A. Waugh, J. J. Heremans, M. B. Santos, X. Liu, and **G. A. Khodaparast**, "Photoinduced spin-polarized current in InSb –based structures, *J. App. Phys.* **106**, 103513 (2009).
- 2-** **Invited Paper: Giti A. Khodaparast**, Matthew Frazier, Rajeev N. Kini, Kanowkan Nontapot, Tetsuya D. Mishima, Michael Santos, Bruce W. Wessels Probe of Coherent and Quantum States in Narrow-Gap Based Semiconductors with Strong Spin-Orbit Coupling Proc. of SPIE **7760**, 77600x-1 (2010)
- 3-** M. Bhowmick, R. N. Kini, K. Nontapot, N. Goel, S. J. Chung, T. D. Mishima, M. B. Santos, **G. A. Khodaparast**, "Probe of Interband Relaxations of Photo-excited and Carriers and Spins in InSb Based Quantum Wells", *Physics Procedia* (3) 1161-1165 (2010).
- 4-** **Invited Paper: Giti A. Khodaparast**, Matthew Frazier, Rajeev N. Kini, Kanowkan Nontapot, Tetsuya D. Mishima, Michael Santos, Bruce W. Wessels, "Probe of Coherent and Quantum States in Narrow-Gap Semiconductors in the presence of strong Spin-Orbit Coupling". Proc. SPIE **7608**, 76080O-1 (2010)
- 5-** M. Bhowmick, T. R. Merritt, K. Nontapot, B. W. Wessels, O. Drachenko, **G. A. Khodaparast**, "Time Resolved Spectroscopy of InMnAs Using Differential Transmission Technique in Mid-Infrared", *Physics Procedia*, (3) 1167-1170 (2010).
- 6-** R. N. Kini , K. Nontapot, **G. A. Khodaparast**, L. J. Guido, R.E. Welser "Time Resolved Measurements of Spin and Carrier Dynamics in InAs Films" , *J. of Applied Physics* **103**, 064318 (2008). *Selected for April 2008 issue of Virtual Journal of Ultrafast Science*.
- 7-** M. Frazier, Nontapot K., Kini R., **Khodaparast G. A.**, Wojtowicz T., Liu X., Furdyna J. K., "Time Resolved Magneto-Optical Studies of Ferromagnetic InMnSb Films", *Appl. Phys. Lett.*, **92**, 061911 (2008).

- 8-** Nontapot K., Kini R. N., Gifford A., Merritt T. R., **Khodaparast G. A.**, Wojtowicz T., Liu X., Furdyna J. K., "Relaxation of Photoinduced Spins and Carriers in Ferromagnetic InMnSb Films, *Appl. Phys. Lett.* **90**, 143109, 2007  
*Selected for the May 2007 issue of Virtual Journal of Ultrafast Science and for the April 16, 2007 issue of Virtual Journal of Nanoscale Science & Technology.*

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### **Interactions/Transitions:**

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Several presentation introduce the research activities pursued in this work including: Two talks at the International Conference on Narrow Gap Semiconductors (July 2005 and July 2007), one talk at the 2008 Electronic Materials Conference, and posters at the Aspen 2007 Winter Conference on Condensed Matter and 2008 Magnetic Excitation in Semiconductors Symposium. Members of Dr. Khodaparast's team gave contributed talks at the APS March meeting [one in 2006, four in 2007, two in 2008, three in 2009, and two in 2010] and presented two posters at the 14<sup>th</sup> International Conference of Narrow Gap Semiconductors in Japan, 2010. One contributed talk was given at the 2010 Electronic Materials , and a poster was presented at The 19th International Conference on the Application of High Magnetic Fields in Semiconductor Physics and Nanotechnology by Prof. Y. Matsuda (Institute of Solid State Physics, Japan), who is a regular collaborator. In addition to several colloquiums, Dr. Khodaparast's presented two recent invited talks in photonic West (Jan. 2010) and SPIE –SPIN-III (August 2010) conferences.

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